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A ^{210}Pb$_{ex}$ mass balance model in cultivated soils in consideration of the radionuclide diffusion*

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The existing ^{210}Pb$_{ex}$ mass balance models for the assessment of cultivated soil erosion are based on an assumption that ^{210}Pb$_{ex}$ is quite evenly mixed within the plough layer. However, the amount of ^{210}Pb$_{ex}$ distributed in the soils below the plough depth, like a downward tail in the lower part of the ^{210}Pb$_{ex}$ profile, has been largely ignored. In fact, after the initial cultivation of undisturbed soils, ^{210}Pb$_{ex}$ will diffuse downward from plough layer to the plough pan layer due to the concentration gradient. Assuming ^{210}Pb$_{ex}$ inventory is constant, the depth distribution in the two layers of the cultivated soils will achieve a steady state after continuous cultivation for 10.37 years, when ^{210}Pb$_{ex}$ is evenly distributed in the soils of the plough layer with an exponential concentration decline with depth in the soils of the plough pan layer, and the ^{210}Pb$_{ex}$ concentration at any depth will be invariable with time. The work reported in this paper attempts to explain the formation of the ^{210}Pb$_{ex}$ tail in the soil profile below the plough depth by theoretical derivation of the ^{210}Pb$_{ex}$ depth distribution process in the two layers of the cultivated soils, propose a ^{210}Pb$_{ex}$ mass balance model considering ^{210}Pb$_{ex}$ diffusion based on the existing model, and discuss the influence of the ^{210}Pb$_{ex}$ tail to the existing model.

Keywords: Cultivated soils, ^{210}Pb$_{ex}$ depth distribution, Diffusion, Mass balance model, Soil erosion

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I. INTRODUCTION

Because of temporal and spatial limitations associated with the application of ^{137}Cs technique to estimating rates of soil erosion and sediment redistribution [1–4], the use of ^{210}Pb$_{ex}$ measurement has attracted increasing attention, as an alternative approach [5–9]. Unlike ^{137}Cs, which is an artificial radionuclide that is released into the environment as a result of atmospheric test of nuclear weapons, ^{210}Pb is a natural product of the ^{238}U decay series, derived from the decay of gaseous ^{222}Rn, the daughter of ^{226}Ra [10, 11]. ^{226}Ra is found naturally in most soils and rocks and the generated ^{210}Pb is in equilibrium with its parent. A small quantity of ^{222}Rn diffuses upward from the soil, introduces ^{210}Pb into the atmosphere and provides an input of this radionuclide to surface soils and sediments, which is not in equilibrium with its parent ^{226}Ra. This fallout component is termed unsupported or excess ^{210}Pb (^{210}Pb$_{ex}$) [12, 13]. Like ^{137}Cs, ^{210}Pb$_{ex}$ is shown to have a strong affinity for soil and sediment particles. Upon reaching the soil surface, ^{210}Pb$_{ex}$ is quickly and strongly adsorbed by the surface soil, and the subsequent redistribution within the landscape will reflect the movement of soil and sediment particles associated with soil erosion and sediment transport processes [14]. Unlike the time-dependent fallout of ^{137}Cs, the atmospheric fallout of ^{210}Pb$_{ex}$ is essentially constant for its natural origin [15], and the ^{210}Pb$_{ex}$ inventory in soils can be invariable because the subsequent fallout provides an input to surface soils that is in equilibrium with the radioactive decay of the existing ^{210}Pb$_{ex}$ in the soils.

In the reported works, the profiles of soils from undisturbed land show that the maximum concentrations of ^{210}Pb$_{ex}$ occur at the surface horizon and decline exponentially with depth (Fig. 1(a), 1(c)), while in the profiles of cultivated soils the ^{210}Pb$_{ex}$ concentration is almost uniform throughout the plough layer as a result of mixing caused by tillage (Fig. 1(b), 1(d)). For the cultivated soils, it is also obvious that an amount of ^{210}Pb$_{ex}$ distributed in the soils below the plough depth, like a downward tail in the lower part of the profile (Fig. 1(b), 1(d)). The ^{210}Pb$_{ex}$ tail in the profile demonstrates that the maximum ^{210}Pb$_{ex}$ concentrations occur at the bottom of plough layer and decline exponentially with depth. However, the amount of ^{210}Pb$_{ex}$ distributed below the plough depth has been largely ignored to date, suggesting that ^{210}Pb$_{ex}$ is quite evenly distributed and restricted to the plough layer. The existing ^{210}Pb$_{ex}$ mass balance models for the assessment of cultivated soil losses have similarly not taken into account the amount of ^{210}Pb$_{ex}$ distributed below the plough depth.

Walling and He [16] developed a mass balance model for estimating erosion rates on cultivated soils from ^{210}Pb$_{ex}$ measurement. Based on this mass balance model, and ignoring the freshly deposited ^{210}Pb$_{ex}$ removed by erosion and influence of the particle size correction factor, Zhang *et al.* proposed a simplified ^{210}Pb$_{ex}$ mass balance model in the steady state for estimating erosion rates on cultivated soils [18], which can be expressed as:

$$I - (\lambda + \frac{\Delta H}{H})A = 0, \tag{1}$$

where A is the ^{210}Pb$_{ex}$ inventory (mBq/cm^2), λ is the decay constant of ^{210}Pb$_{ex}$ (/yr), ΔH is the erosion rate (cm/yr) and H is the plough depth (cm).

The ^{210}Pb$_{ex}$ loss proportion (l) relative to the local reference inventory (A_{ref}, in mBq/cm^2) for an eroding site can be

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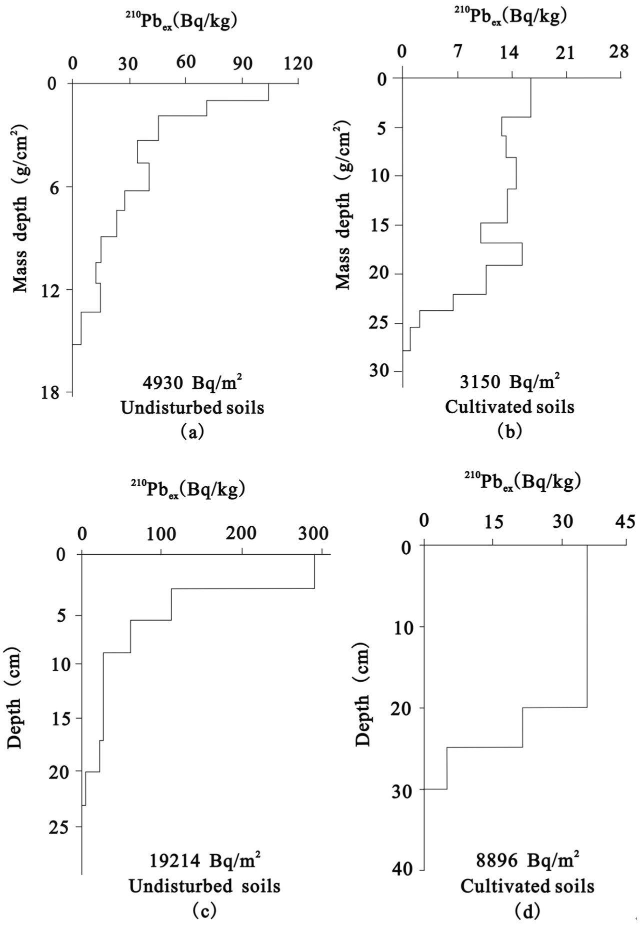

Fig. 1. Typical $^{210}Pb_{ex}$ profiles in undisturbed and cultivated soils. (a), (b), Moorlake catchment, UK [16]; (c), (d), Sichuan Hilly Basin, China [17].

expressed as:

$$l = [(A_{\rm ref} - A)/A_{\rm ref}] \times 100\%. \tag{2}$$

Assuming the $^{210}Pb_{ex}$ deposition flux is constant, the erosion rate can be calculated as Eq. (3) according to the relationship between the deposition flux and the reference inventory ($I = \lambda A_{\rm ref}$).

$$\Delta H = \frac{\lambda H l}{1 - l}. \tag{3}$$

So, the existing mass balance models (existing model for short) are based on the assumption that $^{210}Pb_{ex}$ is quite evenly distributed within the plough depth. Nevertheless, neglecting $^{210}Pb_{ex}$ distributed below the plough depth, more or less, would consequentially influence the application of existing model to soil erosion estimation. So, it is needed to know the influence level, and whether or not the existing model is applicable to soil erosion estimation as usual. In this paper, we attempt to explain the formation of $^{210}Pb_{ex}$ tail in the lower part of profile by theoretical derivation of the $^{210}Pb_{ex}$ depth distribution process in the plough and plough pan layers of the cultivated soils, propose a $^{210}Pb_{ex}$ mass balance model in consideration of the radionuclide diffusion (revision model for short), and discuss the influence of the $^{210}Pb_{ex}$ distributed below the plough depth on the application of existing model to erosion estimation.

II. $^{210}PB_{EX}$ DISTRIBUTION PROCESSES IN CULTIVATED SOILS

A. Mechanism and process analysis

In fact, $^{210}Pb_{ex}$ is quite evenly mixed by tillage within the plough layer, and there is little $^{210}Pb_{ex}$ distributed below the plough depth at the beginning of cultivation, because the plough depth is usually no less than the measured depth range of $^{210}Pb_{ex}$ distribution in the undisturbed soils before tillage. However, this initial depth distribution state is unsteady, and the radionuclide moves downward from the plough layer to the plough pan layer. Radionuclide downward movements in soil include diffusion and migration processes [19]. $^{210}Pb_{ex}$ diffusion in soil is the process of radionuclide movement that is caused by the concentration gradient in the ionic or molecular form, which means that the radionuclide is only able to diffuse from a high concentration to a low concentration. In $^{210}Pb_{ex}$ migration in soil, its downward movement is caused by transportation via media, such as adsorbed solid particles and dissolved fluids, which means that the migration is independent of the concentration gradient, and the radionuclide is able to move either from a high concentration to a low concentration or vice versa. For a soil profile in which the maximum radionuclide occurs at the top horizon and declines monotonically with depth, diffusion can be considered the predominant radionuclide movement process over the profile, while the radionuclide migration processes can be neglected [20–24]. Thus, it can be assumed that the diffusion process dominates the formation of the $^{210}Pb_{ex}$ tail in the profile of the plough pan layer.

At the beginning of cultivation, $^{210}Pb_{ex}$ is quite evenly distributed within the plough layer; after the initial cultivation, the $^{210}Pb_{ex}$ diffuses from the plough layer to the plough pan layer due to the concentration gradient; the $^{210}Pb_{ex}$ inventory of the plough layer decreases while that of plough pan layer increases; and the $^{210}Pb_{ex}$ tail in the profile of the cultivated soils progressively comes into being closely below the interface between the two layers (Fig. 1(b), 1(d)). Assuming the $^{210}Pb_{ex}$ inventory is constant, the $^{210}Pb_{ex}$ concentration at any soil depth will be invariable after a period of diffusion, and the $^{210}Pb_{ex}$ depth distribution in the two layers of cultivated soils will achieve a steady state.

B. The depth distribution in the unsteady state

Assuming no soil erosion or deposition occurs, when $^{210}Pb_{ex}$ is evenly mixed within the plough layer by the initial cultivation of undisturbed soils. Subsequently, the $^{210}Pb_{ex}$

starts moving from the plough layer to the plough pan layer. Taking the diffusion process into account, the ^{210}Pb$_{\rm ex}$ movement in the soils of plough pan layer follows Fick's second law of diffusion [25], which is:

$$D\frac{\partial}{\partial z}\left(\frac{\partial C_{\rm pp}(z,t)}{\partial z}\right)-\frac{\partial C_{\rm pp}(z,t)}{\partial t}-\lambda C_{\rm pp}(z,t)=0, \quad (4)$$

where D is the effective diffusion coefficient (cm^2/yr), $C_{\rm pp}$ is the ^{210}Pb$_{\rm ex}$ concentration in the soils of the plough pan layer (mBq/cm^3), z is the depth downward from the interface between the plough and plough pan layers (cm), and t is the time elapsed since ^{210}Pb$_{\rm ex}$ was well mixed within the plough layer (yr). When $z\to 0$, the ^{210}Pb$_{\rm ex}$ concentration of the interface soils is equal to the value of the ^{210}Pb$_{\rm ex}$ concentration at any depth of the plough layer.

By diffusion, the total ^{210}Pb$_{\rm ex}$ (A, in mBq/cm^2) is redistributed between the plough and plough pan layers, which means:

$$A=A_{\rm p}+A_{\rm pp}, \quad (5)$$

where $A_{\rm p}$ and $A_{\rm pp}$ are the ^{210}Pb$_{\rm ex}$ inventory of the plough layer and plough pan layer, respectively.

The ^{210}Pb$_{\rm ex}$ inventory in the soils of the plough layer can be calculated as:

$$A_{\rm p}=HC_{\rm p}. \quad (6)$$

Assuming the plough pan layer can be treated as a semi-infinite homogeneous porous medium, the process of ^{210}Pb$_{\rm ex}$ diffusion from the plough layer to the plough pan layer can be considered as the infinite thin layer radionuclide resource diffusing to a one-dimensional semi-infinite medium. Thus, according to the Gauss solution of the Fick's second law [26], by combining Eqs. (4), (5) and (6), the ^{210}Pb$_{\rm ex}$ concentration variation with time in the soils of the plough layer can be expressed as:

$$C_{\rm p}(t)=A/[H+(\pi Dt)^{1/2}]. \quad (7)$$

And the variation of the ^{210}Pb$_{\rm ex}$ concentration with time and depth in the soils of the plough pan layer can be expressed as:

$$C_{\rm pp}(z,t)=\frac{A}{H+\sqrt{\pi Dt}}e^{-\frac{z^2}{4Dt}}. \quad (8)$$

By integrating the two sides of Eq. (8) over depth z, the proportion of the ^{210}Pb$_{\rm ex}$ inventory of the plough pan layer to the total ^{210}Pb$_{\rm ex}$ inventory in the cultivated soil, namely the ^{210}Pb$_{\rm ex}$ diffusion proportion, can be expressed as:

$$\frac{A_{\rm pp}(t)}{A}=\frac{\sqrt{\pi Dt}}{H+\sqrt{\pi Dt}}\times 100\%. \quad (9)$$

The above theoretical derivation, namely Eqs. (4)–(9), can also apply to the ^{137}Cs depth distribution process in cultivated soils. However, unlike ^{137}Cs, the ^{210}Pb$_{\rm ex}$ depth distribution in the two layers of the cultivated soils finally achieves a steady state after a period of diffusion, assuming the ^{210}Pb$_{\rm ex}$ deposition is constant over time. Before the steady state, the ^{210}Pb$_{\rm ex}$ concentration at any depth of the two layers varies with time. If the effective diffusion coefficient D is known (which can be obtained by experiments), Eq. (7) can be used to simulate the ^{210}Pb$_{\rm ex}$ concentration variation with time in the soils of the plough layer, and Eq. (8) can be used to simulate the ^{210}Pb$_{\rm ex}$ concentration variation with time and depth in the plough pan layer.

C. The depth distribution in the steady state

After achieving a steady state, the ^{210}Pb$_{\rm ex}$ concentration at any depth of the cultivated soils is invariable with time, viz. $\partial C_{\rm pp}(z,t)/\partial t=0$, and Eq. (4) can be written as:

$$D\frac{\partial}{\partial z}\left(\frac{\partial C_{\rm pp}(z)}{\partial z}\right)-\lambda C_{\rm pp}(z)=0. \quad (10)$$

When $z\to 0$, $C_{\rm pp(0)}$ is the ^{210}Pb$_{\rm ex}$ concentration in the interface soils between the plough and the plough pan layer, which represents the ^{210}Pb$_{\rm ex}$ concentration at any depth of the plough layer or the ^{210}Pb$_{\rm ex}$ concentration in the top soils of the plough pan layer. Based on this boundary condition, Eq. (10) can be solved as follows:

$$C_{\rm pp}(z)=C_{\rm p}e^{-(\lambda/D)^{\frac{1}{2}}z}. \quad (11)$$

Integrating the two sides of Eq. (11) over the depth (z), the following can be derived:

$$A_{\rm pp}(t)=C_{\rm p}(D/\lambda)^{1/2}. \quad (12)$$

By combining Eqs. (5), (6), (11) and (12), the ^{210}Pb$_{\rm ex}$ concentration in the soils of the plough layer in the steady state can be expressed as:

$$C_{\rm p}=\frac{A}{H+\sqrt{D/\lambda}}. \quad (13)$$

And the variation of the ^{210}Pb$_{\rm ex}$ concentration with depth in the plough pan layer in the steady state can be expressed as:

$$C_{\rm pp}(z)=\frac{A}{H+\sqrt{D/\lambda}}e^{-(D/\lambda)^{\frac{1}{2}}z}. \quad (14)$$

By combining Eqs. (9), (12) and (13), the time elapsed from initial cultivation to the steady state can be calculated as:

$$t_{\rm steady}=1/(\lambda\pi)=10.37\,\text{yr}. \quad (15)$$

And the diffusion proportion in the steady state can be expressed as:

$$\begin{aligned}k&=\frac{A_{\rm pp}}{A}\times 100\%\\&=\frac{\sqrt{D/\lambda}}{H+\sqrt{D/\lambda}}\times 100\%,\end{aligned} \quad (16)$$

where k is the diffusion proportion in the steady state, which can be obtained by experiments.

From Eq. (16), the effective diffusion coefficient of ^{210}Pb$_{ex}$ is:

$$D = \lambda \left(\frac{kH}{1-k} \right)^2 . \quad (17)$$

Equations (13) and (14) can be used to simulate the ^{210}Pb$_{ex}$ depth distribution in the steady state in the soil profile for the two layers of the cultivated land. A basic assumption of this theoretical derivation is that no erosion or deposition occurs. For an eroding soil, the ^{210}Pb$_{ex}$ inventory would be invariable assuming the erosion rate is constant for over 100 years. Then the ^{210}Pb$_{ex}$ depth distribution will also be in steady state, and Eqs. (10)–(17) can be applicable as usual.

True, ^{210}Pb$_{ex}$ downward diffusion below the plough depth might be partially offset by soil erosion at the surface, causing the base of the plough layer to be displaced downwards, and some ^{210}Pb$_{ex}$ that has diffused downwards would return to the plough layer. However, the ^{210}Pb$_{ex}$ distributed below the plough depth, more or less, is a matter of fact. In the cultivated purple soil of the Sichuan Hilly Basin in China (Fig. 1(d)), for example, ca. 37% of the downwards diffused ^{210}Pb$_{ex}$, according to Eq. (8), returned to the plough layer at the first year of diffusion, assuming ^{210}Pb$_{ex}$ is initially quite evenly distributed within the plough depth and the soil erosion rate is 0.5 cm/yr (the real erosion rates are usually less than this value [17]). In a word, ^{210}Pb$_{ex}$ diffusion in cultivated soils is a matter of fact in spite of the usual erosion. And the proportion would decrease with year and eventually tend to constant. Then, the ^{210}Pb$_{ex}$ distribution would achieve the steady state with a ^{210}Pb$_{ex}$ tail in the profile below the plough depth.

D. The simulated ^{210}Pb$_{ex}$ diffusion process in a soil core from the Yimeng mountains area

To know the ^{210}Pb$_{ex}$ depth distribution in cultivated soil in reality, a typical ^{210}Pb$_{ex}$ profile for a core from the cultivated cinnamon soils of the Yimeng mountains area in China was introduced in this study [27], based on which the ^{210}Pb$_{ex}$ diffusion process could be simulated for the core. The Yimeng Mountains area (ca. 17 180 km^2) is located in the central south of Shandong Province, China (34°22′ to 36°13′N, 117°24′ to 119°11′E), with elevations within the range of 150–1165 m. The area belongs to the continental monsoon climate area of the warm temperate zone, and the cinnamon soils developed from limestone weathering crusts are wildly distributed. The core was collected from the fields situated in flat areas at the top of ploughed fields, where the disturbance except for cultivation over the past 100 years can be negligible. The profile (Fig. 2(a)) reflects the steady state of ^{210}Pb$_{ex}$ depth distribution because the soils have not been disturbed except for cultivation in nearly ca. 100 years. From the profile and the field surveys it can be determined that, the plough depth (H) is ca. 20 cm because the ^{210}Pb$_{ex}$ concentrations in soils of the upper ∼ 20 cm are relatively uniform, suggesting the ^{210}Pb$_{ex}$ is mixed by tillage within the uppermost depth of ∼ 20 cm, which is also in agreement with our field surveys; the ^{210}Pb$_{ex}$ diffusion proportion (k) is 32%, which is the ratio of the ^{210}Pb$_{ex}$ inventory below the plough depth to the total inventory in the profile; the effective diffusion coefficient (D) is 2.72 cm^2/yr, which can be calculated using Eq. (17).

Equations (7) and (8) can simulate the ^{210}Pb$_{ex}$ redistribution process in the plough and plough pan layer of cinnamon soils after the initial cultivation before the steady state (e.g. t = 2, 4, 6, and 8 yr); Eqs. (13) and (14) can simulate the stable ^{210}Pb$_{ex}$ depth distribution in the two layers of the cultivated cinnamon soils ($t \geq 10.37$ yr) (Fig. 2(b)).

III. MASS BALANCE MODEL IN CONSIDERATION OF THE DIFFUSION

A. Model derivation

Considering ^{210}Pb$_{ex}$ depth distribution in the cultivated land in view of the radionuclide diffusion from the plough to the plough pan layer, Eq. (1) can be rewritten as:

$$I - \lambda A - \Delta H A_{\mathrm{p}}/H = 0. \quad (18)$$

By combining Eqs. (2), (6), (13), (17) and (18), the erosion rate of the cultivated soil can be expressed as Eq. (19) based on the relationship between the ^{210}Pb$_{ex}$ deposition flux and the reference inventory ($I = \lambda A_{\mathrm{ref}}$).

$$\Delta H = \frac{\lambda H l}{(1-k)(1-l)}. \quad (19)$$

Therefore, considering the ^{210}Pb$_{ex}$ diffusion from the plough layer to the plough pan layer, the erosion rates can be estimated from Eq. (19). By ignoring the influence ^{210}Pb$_{ex}$ diffusion ($k = 0$), Eq. (19) can be reduced to Eq. (3).

B. Comparison of the revision and existing models

As above, the difference between the revision and existing model lies in whether to take into account the amount of ^{210}Pb$_{ex}$ distributed below the plough depth. To compare the two models, let ΔH_1 and ΔH_2 be the erosion rates estimated using Eqs. (3) and (19), respectively, and we have:

$$\Delta H_1 = \lambda H l/(1-l), \quad (20)$$

$$\Delta H_2 = \lambda H l/[(1-k)(1-l)]. \quad (21)$$

It is clear that $\Delta H_1 < \Delta H_2$, and the difference ratio (d) of ΔH_1 and ΔH_2 is:

$$d = (\Delta H_2 - \Delta H_1)/\Delta H_2, \quad (22)$$

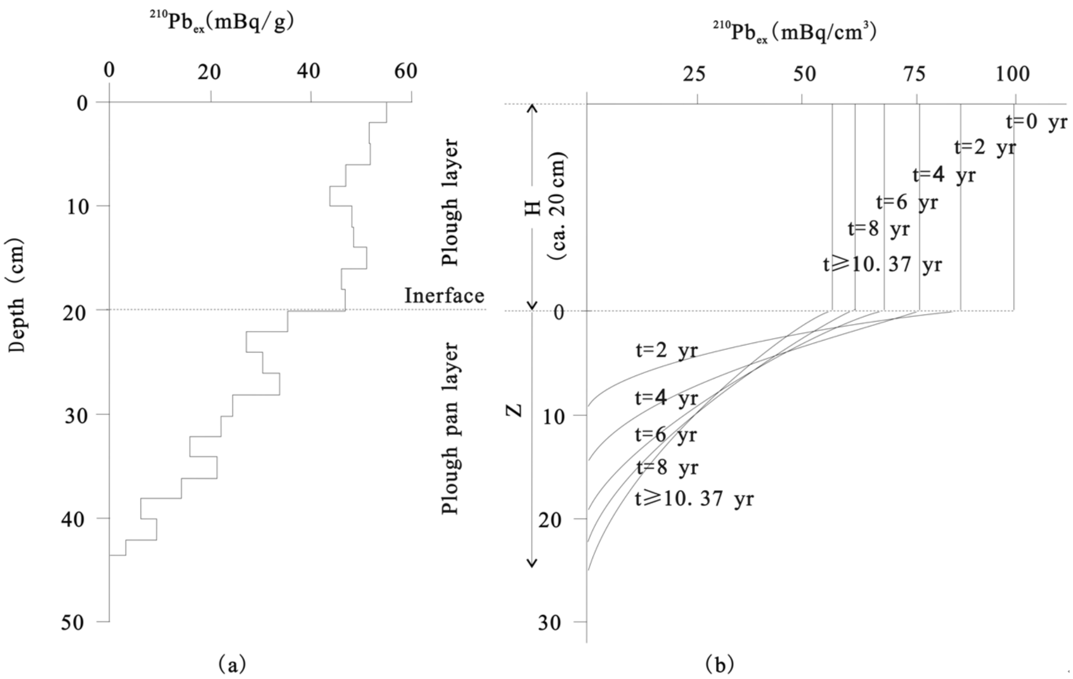

Fig. 2. (a) The typical ^{210}Pb$_{ex}$ profile for a core from the cultivated cinnamon soils of the Yimeng mountains area [27]; (b) The simulated ^{210}Pb$_{ex}$ redistribution process in the core after the initial cultivation.

By combining Eqs. (20)–(22), one has:

$$d = k. \tag{23}$$

So, the difference ratio can be represented by the ^{210}Pb$_{ex}$ diffusion proportion in the steady state of the ^{210}Pb$_{ex}$ depth distribution, and the difference increases with the proportion of diffusion.

If the ^{210}Pb$_{ex}$ diffusion proportion (k) and loss proportion (l) are known, Eq. (19) can be used to estimate the erosion rates. These parameters can be derived from the experiments. Because the k is determined by the ^{210}Pb$_{ex}$ profile measurement, the accuracy of k is related to the accuracy of experiment, e.g. the study site selection, sampling method, the sampling section increments, laboratory measurement and etc.. Moreover, the recognition of the plough depth, according to the shape of the profile in conjunction with field survey, is also important to the accuracy of k.

IV. DISCUSSION AND CONCLUSION

As above, the erosion rates estimated using the revision model are higher than those estimated using the existing model by comparison. In other words, the existing model underestimates the erosion rate of the cultivated soils to some extent because of ignoring the amount of ^{210}Pb$_{ex}$ distributed below the plough depth. Nevertheless, the existing model involves the superiorities including compact form, fewer parameters, and convenience for application. In contrast, the revision model involves the limitations including complicated form and more parameters for application. Eq. (23) suggests that the underestimation of soil erosion from the existing model is as well less than 10% when the ^{210}Pb$_{ex}$ diffusion proportion (k) is lower than 10%, which could be negligible. In other words, the existing model is as usual applicable to soil erosion estimation when the diffusion proportion is lower than 10%. When the diffusion proportion ranges between 10%–15%, the existing model is also suitable if the underestimation is acceptable in some practical applications. However, the underestimation could not be neglected when the diffusion proportion is greater than 15%, and the revision model would be more applicable to erosion estimation than the existing model. No matter what the diffusion proportion is, the revision model is universal because the existing model is merely a case of the revision model (when $k = 0$). In a word, the application scope of revision model is universal, especially for the locations where the diffusion proportions in the cores are greater than 15%.

Equation (17) indicates that the diffusion proportion is related to the diffusion coefficient (D) and the plough depth (H), viz. the diffusion proportion increases with the diffusion coefficient while decreases with the plough depth. The diffusion coefficients and the plough depths vary in different areas with different soil types and tillage systems. In the reported works, the diffusion proportions are usually less than 10%, e.g. a typical range of 5%–7% at the study sites in UK [16, 19] and southern Italy [28, 29]. In these cases, the diffusion proportion can be negligible, and the existing model is applicable as before. For cultivated brown and cinnamon soils of the Yimeng mountains area in China, however, the diffusion proportions are greater than 15% [27], so the revi-

sion model is more applicable to erosion estimation by comparison.

Neglecting the $^{210}Pb_{ex}$ below the plough depth, as above, will always underestimate soil erosion, nevertheless, it can also lead to an overestimation in some cases, e.g., erosion of uncultivated soils might be overestimated when the reference site is selected in a cultivated soil if the $^{210}Pb_{ex}$ below the plough depth is neglected, viz. the sampling depth is restricted to the plough depth.

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